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MEASUREMENT OF THE LINE REVERSAL
TEMPERATURE OF OH IN $\text{CH}_4/\text{N}_2\text{O}$ FLAMES

William R. Anderson

January 1981



US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
BALLISTIC RESEARCH LABORATORY
ABERDEEN PROVING GROUND, MARYLAND

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER TECHNICAL REPORT ARBRL-TR-02280	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) MEASUREMENT OF THE LINE REVERSAL TEMPERATURE OF OH IN CH ₄ /N ₂ O FLAMES		5. TYPE OF REPORT & PERIOD COVERED Technical Report
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) William R. Anderson		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS U.S. Army Armament Research & Development Command U.S. Army Ballistic Research Laboratory ATTN: DRDAR-BLI Aberdeen Proving Ground, MD 21005		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 1L161102AH43
11. CONTROLLING OFFICE NAME AND ADDRESS U.S. Army Armament Research & Development Command U.S. Army Ballistic Research Laboratory ATTN: DRDAR-BLI Aberdeen Proving Ground, MD 21005		12. REPORT DATE JANUARY 1981
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		13. NUMBER OF PAGES 24
		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release, distribution unlimited		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Band reversal Line reversal Flame temperature OH radical		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) (clt) The reversal technique is universally accepted by the combustion community as an accurate method for flame-temperature measurements. This technique has generally involved the use of Na seeded in flames. However, for many burner types seeding is difficult. This work describes band-reversal measurements using OH, which occurs naturally in all flames containing oxygen and hydrogen. The theory is discussed as well as use of the technique in the burnt gas region of CH ₄ /N ₂ O flames. The experimental results are compared with laser-excited		
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20. ABSTRACT (cont'd)

fluorescence temperature measurements also performed in this laboratory. Excellent agreement was achieved.

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I. INTRODUCTION

Temperature measurement is of vital interest in studies of flames. It is necessary for determination of number densities of species in flames from spectroscopic data. It is also useful for comparison with the results of detailed modeling calculations. One of the better accepted spectroscopic techniques for the measurement of flame temperature is the reversal method,¹ which has generally been applied using the sodium D lines. The technique is then termed line reversal. For certain burner types, however, Na seeding is difficult or impossible. In these cases it is wise to look for a transition involving one of the flame species. This paper describes application of reversal to the (0,0) R bandhead of the OH $A^2\Sigma^+ \rightarrow X^2\Pi$ system. The technique is best termed band reversal.

Reversal techniques do not provide the spatial resolution of laser based techniques, and measurements therefore cannot be made in the flame front by reversal. Such measurements, however, can be easily performed in the burnt gas region where the temperature gradients are not large. The results can be used for comparative purposes with those of the laser based methods.

This paper is written as an introduction to line reversal to point out some subtleties in what is conceptually a very simple experiment. First the theory behind the technique is discussed, then the experimental apparatus is discussed and, finally, results of reversal measurements are presented and compared with laser excited fluorescence measurements.

II. THEORY

Consider a gas inclosed by a blackbody in equilibrium at temperature T. Suppose that only radiative heat transfer is possible between the walls of the blackbody and the gas. The spectral intensity in the frequency range ν to $\nu + d\nu$ is isotropic and is given by²

¹A. G. Gaydon and H. G. Wolfhard, Flames: Their Structure, Radiation and Temperature, (Chapman and Hall, LTD., London, 1970).

²Y. B. Zel'dovich and Y. P. Raizer, Physics of Shock Waves and High Temperature Hydrodynamic Phenomena, (Academic, New York, 1966, Vol. 1, Chapter 2; S. S. Penner, Quantitative Spectroscopy and Gas Emissivities, (Addison-Wesley, Inc., Reading, Mass., 1959); L. D. Landau and E. M. Lifshitz, Statistical Physics (Addison-Wesley, Inc., Reading, Mass, 1958).

$$I_v dv = \frac{2hv^3}{c^2} \frac{1}{\exp(hv/kT) - 1} dv \quad (1)$$

where h is Planck's constant, c the speed of light and k Boltzmann's constant. Since some confusion might easily result, it is worthwhile to discuss the physical meaning of I_v and its relation to other observables in radiation theory. The quantity I_v is the amount of radiant energy in the spectral interval dv passing a unit area in space in unit time. The propagation vectors of I_v are restricted to lie within a solid angle $d\Omega$ about the normal to the unit surface area. Typical units for I_v are therefore erg/cm² sec Hz ster. Two other radiant energy observables are typically discussed. They are the spectral energy density, ρ_v , and the intensity of parallel radiation, I_{vp} . The energy density is essentially hv times the number of photons in the range dv present in a unit volume at a given instant in time (for purposes of the present study, all observables are assumed to be time independent). The intensity of parallel radiation is defined for parallel radiation, as, for example, from lasers, and is the amount of radiant energy in the spectral interval dv passing a unit area perpendicular to the direction of propagation in unit time. For isotropic radiation, $\rho_v = (4\pi/c)I_v$. For parallel radiation, $\rho_v = (1/c)I_{vp}$. Typical units for ρ_v are thus erg/cm³Hz and for I_{vp} are erg/cm² Hz sec. Care must be taken to avoid error by a factor 4π if intensity is used, especially in calculations of absorption or emission rates.

The steradiancy² of an emitting surface is defined as the amount of radiant energy in unit frequency interval emitted per unit time from a unit area of the surface into unit solid angle. The intensity at a surface parallel to the emitter surface and placed along the normal to the emitter, but a great distance away, is equal to the steradiancy normal to the emitter. Therefore, for a blackbody emitter surface, the steradiancy normal to the surface is also given by Eq. 1.*

Let us now consider the gas at equilibrium with the blackbody enclosure. Suppose a transition between lower level 1 and upper level 2 of energy hv is possible for the gas. The ratio of the number densities in the two states is then given by the familiar Boltzmann law:

$$N_2/N_1 = (g_2/g_1)\exp(-hv/kT) \quad (2)$$

* The steradiancy at an angle θ to the normal of the emitter surface drops by the factor $\cos \theta$ (see Ref. 1 and 2). One must take care to use only radiation from a small solid angle normal to the surface of calibrated emitters to avoid error.

where N_i is number density and g_i the level degeneracy. Radiative transfer occurs via three processes: absorption from the ground state, stimulated emission from the excited state and spontaneous emission from the excited state. The rate of absorption is given by $B_{12} I_\nu N_1$, stimulated emission by $B_{21} I_\nu N_2$ and spontaneous emission by $A_{21} N_2$, where A_{21} , B_{12} and B_{21} are the Einstein coefficients expressed in terms of spectral intensity.^{2,3} At equilibrium, the rate of absorption is equal to that of emission, assuming nonradiative energy transfer is negligible, and then

$$B_{12} I_\nu N_1 = B_{21} I_\nu N_2 + A_{21} N_2 . \quad (3)$$

By substituting Eq. 2 into 3 and solving for I_ν (after rearrangement to the form of Eq. 1), one finds²

$$B_{21} = c^2 A_{21} / 2h\nu^3 \quad (4a)$$

and

$$B_{12} = (g_2/g_1) B_{21} . \quad (4b)$$

Eqs. 4a and b were originally derived by Einstein as described above.* Rearranging Eq. 3 one finds

$$(B_{12} N_1 - B_{21} N_2) I_\nu = A_{21} N_2 . \quad (5)$$

What happens if the blackbody and gas are not in equilibrium? Suppose the blackbody temperature, T_b , is lower than the gas temperature, T_g . Since I_ν is smaller at the lower temperature, the spontaneous emission is larger than the net absorption. There will thus be a net emission at ν . Similarly, if $T_g < T_b$, there will be a net absorption at ν . If $T_g = T_b$, there is no absorption or emission.

³A. C. G. Mitchell and M. W. Zemansky, Resonance Radiation and Excited Atoms, (Cambridge University Press, 1934) p. 92-5.

*Einstein's original derivation of the relation was in terms of energy density, ρ_ν . One must be extremely careful in rate calculations that the observable one is using (ρ_ν , I_ν or $I_{\nu p}$) is appropriate for the dimensions of B_{12} . An error by a hidden factor of 4π can easily result otherwise.

A blackbody as described above is impractical for experimental work. If one places a blackbody emitter and a detector subtending the same solid angle, $d\Omega$, on either side of a gas confined to the volume V , as shown in Figure 1, one may analyze the situation for practical experiments. Since the spontaneous emission is isotropic, the rate of spontaneous emission into the detector is $A_2 N_2 V d\Omega / 4\pi$. The stimulated terms in Eq. 5 are affected by an equivalent factor since the solid angles for source and detector are equivalent. Thus, an experiment can be run without integrating over 4π ster.* The solid angle of the detector must be the limiting aperture, not the emitter. Otherwise, the emission of some of the flame gases, with no background irradiation, will enter the detector, causing erroneous measurements.

Let us now consider the most common emission source used in reversal work, the tungsten ribbon lamp. Such a lamp contains a strip of tungsten which is heated by a constant current source. Light emitted from the filament may pass through a pyrex window. However, this work necessitated use of a lamp with a fused silica window to pass ultraviolet radiation. The emissivity of tungsten is not unity so that Eq. 1 does not apply. Instead, the steradiancy is multiplied by an emissivity factor, E . Converting to unit wavelength interval $d\lambda$, since the lamp used in this work was calibrated in such units, and remembering $|dv| = cd\lambda/\lambda^2$, one finds the intensity normal to the surface is

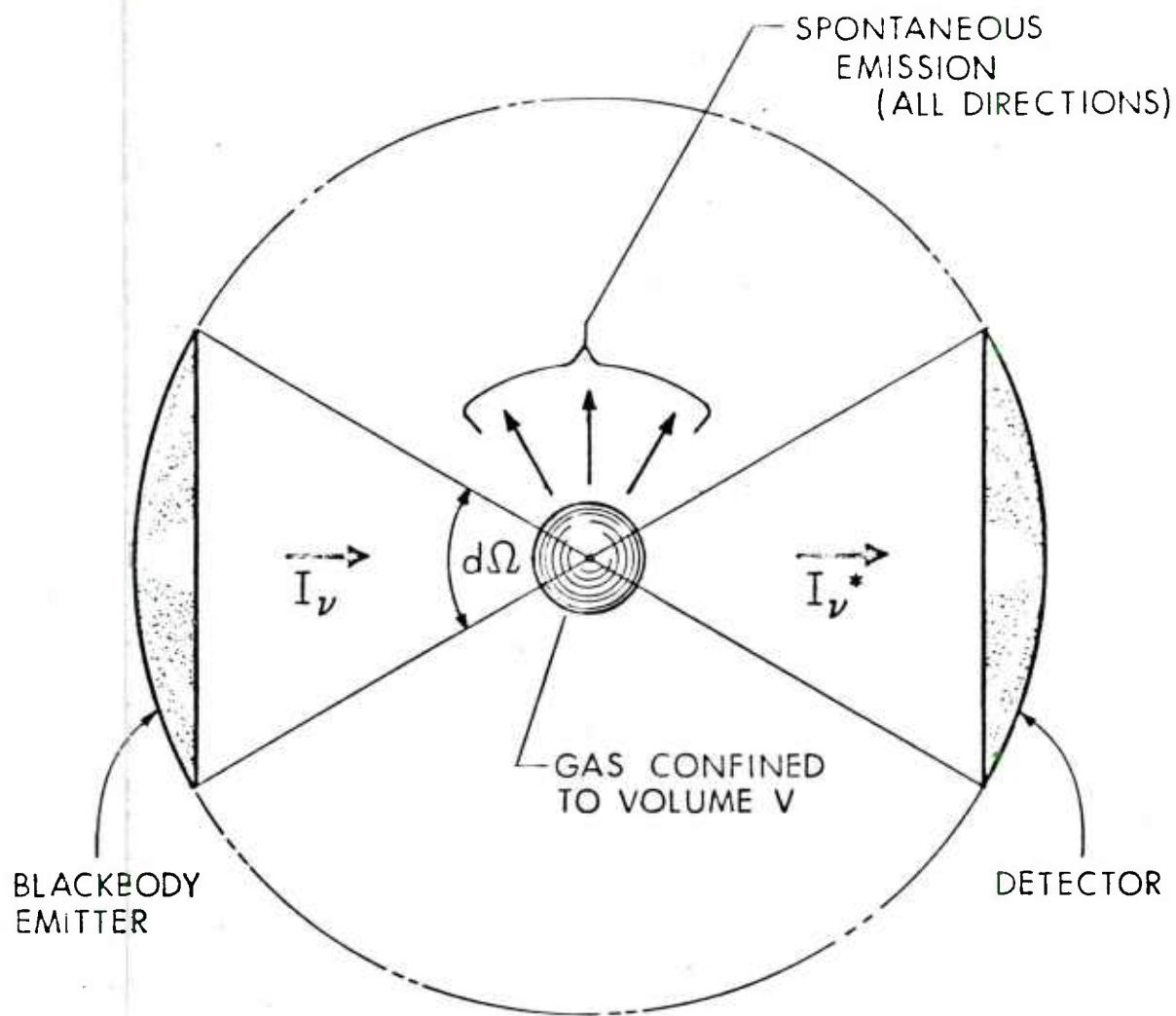
$$I_\lambda d\lambda = \frac{2hc^2}{\lambda^5} \frac{E(\lambda, T)}{\exp(hc/\lambda kT) - 1} d\lambda \quad (6)$$

where T is the temperature of the filament. Tables of emissivity for tungsten may be found in ref. 1, if needed. However, the method described in this work requires usage of Eq. 6 only for a hypothetical blackbody emitter which has $E = 1$.

III. EXPERIMENTAL

A diagram of the apparatus used in this work is shown in Figure 2. The standard lamp is a tungsten ribbon lamp with a fused silica exit window which is calibrated for steradiancy normal to the surface vs.

*In this design, the spontaneous emission loss is not compensated by a surrounding blackbody enclosure. Collisional excitation and quenching usually dominate the energy partitioning. However at sufficiently low pressures, as discussed in ref. 1, the radiative losses may appreciably lower the temperature of the electronic modes. This phenomenon can result in measured reversal temperatures which are quite low compared to the translational temperature.



$$I_\nu^* = I_\nu - \text{ABSORPTION} + \text{STIMULATED EMISSION} + \text{SPONTANEOUS EMISSION OCCURRING IN } d\Omega$$

Figure 1. Gedanken reversal experiment for an emitter-detector combination of solid angle $d\Omega$. In a real experiment, the gas in volume V is replaced by a flame and the emitter by a lens. The blackbody emitter could equally well be represented by a point blackbody emitter at the center of the sphere.

REVERSAL APPARATUS

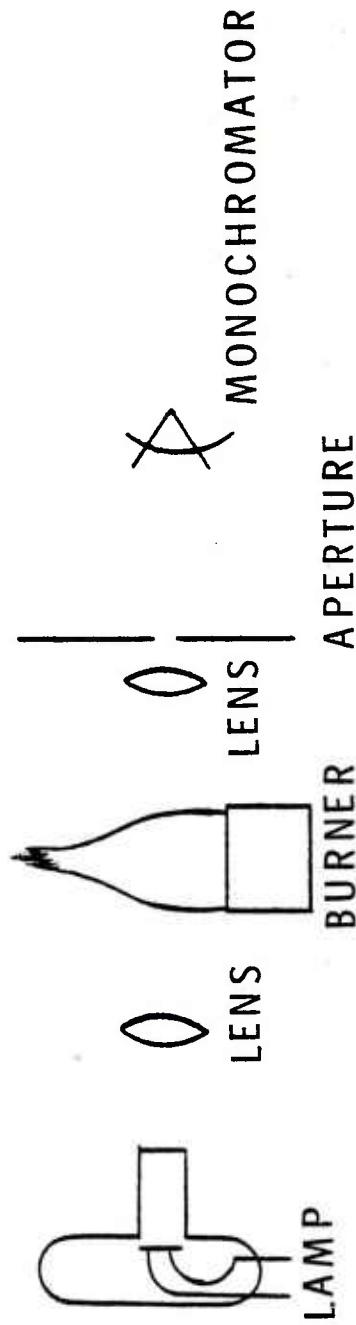


Figure 2. Diagram of Reversal Apparatus. The lamp is a tungsten ribbon lamp with a fused silica window. The lenses are also made of fused silica. The apparatus is adjustable. The apparatus is set up for 1:1 imaging of the tungsten filament at the center of the flame and at the monochromator slits.

wavelength at a constant current of 35 amp (Eppley Laboratory, Inc.). A similar working lamp is used for most measurements and later calibrated against the standard lamp. Light from the lamp is 1:1 imaged into the flame by a quartz lens. Thus, the intensity at the flame is equal to the lamp steradiancy (neglecting lens losses). The lamp radiation and flame emission are 1:1 imaged on the slits of a monochromator. An aperture stop limits the acceptance angle of the monochromator so that it views equal solid angles of the flame and background radiation. The aperture must be small enough to restrict the background emission to that portion which is nearly normal to the filament.

Care must be taken to avoid chromatic aberration of the lenses. Variation in the index of refraction with wavelength causes the focal length of the lenses used to change from 12.70 cm in the visible to 12.07 cm at 3070 Å, as calculated using the lensmakers equation. Ignorance of this change can introduce a 100-200 K systematic error into the results.

As previously stated, the flames used in this work were CH₄/N₂O flames. The flames were produced by a water-cooled, sintered bronze flat-flame type burner of 6.4 cm diameter. Capillary flow regulators provided supply gases to the burner. The absolute flow rates were accurate to within 1%.⁴

A 0.3-m Heath monochromator was used in this work. A 100-micron slitwidth was used yielding a bandpass of ~ 3 Å FWHM. Output from the monochromator was detected by an EMI 9558QA photomultiplier (PMT). Current from the PMT was monitored by a picoammeter. Hard copy of the picoammeter output was obtained on a strip-chart recorder.

The transmission of the first lens after the lamp was determined in the following manner. The PMT response was determined with the apparatus set up as shown in Figure 2 with the flame off. Then the lens and burner were removed. The lamp was moved to the point where the tungsten filament was above the center of the original burner position, i.e., at the original imaging point. The ratio of PMT responses yields the transmission. Of course, the lamp supply current, slitwidth and wavelength observed must be held constant.

In a typical temperature run, the monochromator was scanned from about 3050 to 3080 Å across the R₁ and R₂ bandheads of the OH (0,0) vibrational band. Figure 3 shows a scan of the flame emission with the lamp turned off. The R₁ and R₂ bandheads appear as a splitting on the peak between 3060 and 3075 Å. The sharp rise at 3060 Å and the splitting of the peak were found to be very useful in temperature runs with low signal to noise ratio. For a given flame, the lamp current is changed and the monochromator is scanned each time until the reversal point is

⁴M. A. Dewilde, "Capillary Flowmeters for Accurate Stable Flows of Gases". BRL Technical Report ARBRL-TR-02230, March 80. (AD #A083874)

OH A \rightarrow X EMISSION IN CH₄/N₂O FLAME

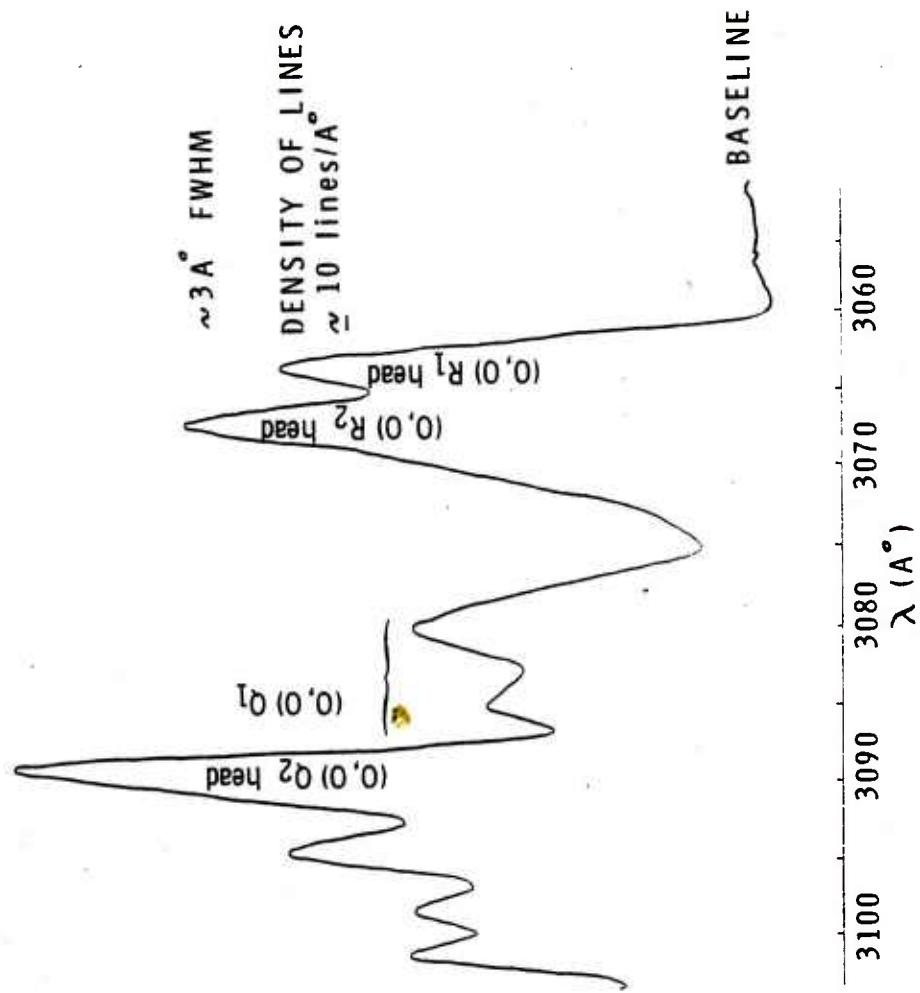


Figure 3. OH Emission Scan in CH₄/N₂O Flame

obtained. A typical set of scans is shown in Figure 4. The graybody curve of the lamp is shown in Figure 4a with the flame off. With the flame on, the OH spectrum shows up, in Figure 4b, as an emission. The splitting of the R₁ and R₂ bandheads may be discerned on the scan. The peak positions are pointed out by arrows on the scan. Though the exact temperatures are not known, the background temperature was lower than, but very close to, the flame temperature. Once the reversal point had been determined, the monochromator was scanned with the lamp current set + 0.5 and - 0.5 amps away from the reversal point to be sure that absorption and emission, respectively, could be discerned. The ± 0.5 amp current setting typically introduced ± 25K into the error limits.

The reversal temperature was determined in the following manner. The monochromator was set at 3070 Å, the approximate center of the OH peak used. The PMT current was then determined for the working lamp at the reversal current and for the standard lamp at its calibration current (35 amps). The steradiancy, S, of the standard lamp was known from its calibration vs. wavelength. The intensity at the reversal point is thus

$$I(3070 \text{ } \overset{\circ}{\text{A}}) = (C_w/C_s)St \quad (7)$$

where C_w is the PMT current for the working lamp at the reversal point, C_s that for the standard lamp at its calibration current and t is the transmission of the lens. The temperature of the flame is then that at which a blackbody (not the tungsten filament!!!) produces the intensity of Eq. 7. This temperature is determined by inserting E = 1 in Eq. 6 and solving for T.

The absorption measured in this work was of the order of 5%. This low signal level made discerning the absorption peak from background noise very difficult. In retrospect, it would probably have been better to use a high resolution monochromator with bandwidth of order 0.05 Å, the approximate bandwidth of a single rotational line of OH in the flame, and use pulse counting techniques. The signal to noise ratio should be much higher and the reversal of individual lines could be examined.

No other species besides OH were used in this study. The primary reason for this restriction was the impracticality of seeding the porous sintered bronze burner with sodium salts or other materials commonly used in reversal work. A scan of emission from the burnt gas region of the CH₄/N₂O flames revealed the presence of minor emissions from NH (3360 - 70 Å) and CN (3880 Å) which probably were too small to yield useful results by this technique. (However, with a higher resolution monochromator they might be used). Strong H₂O bands were observed at 6930 and 7165 Å. The first has a sufficiently sharp bandhead that it might yield good results, but neither was tried in this study.

OH BAND REVERSAL

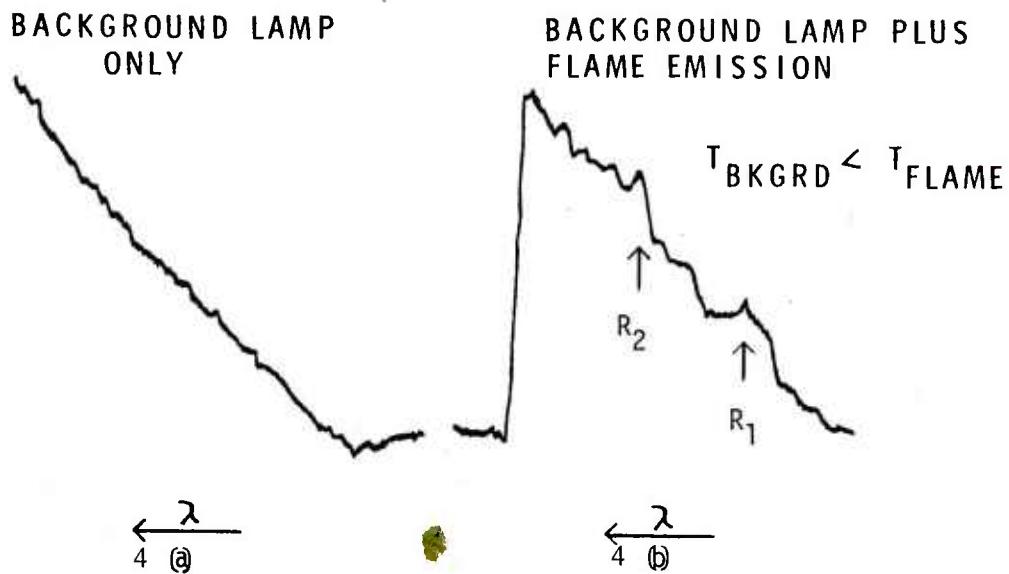


Figure 4. Scan of lamp plus OH Emission. The R₁ and R₂ bandheads of Figure 3 are pointed out in Figure 4b.

The OH reversal temperature was determined in the burnt gas region of stoichiometric, slightly lean and slightly rich CH₄/N₂O flames. The results are presented in the next section and compared with the results of laser-excited fluorescence measurements.

IV. DISCUSSION

The flame temperature was measured in the burnt gas region of several CH₄/N₂O flames by line reversal and at several heights by OH laser-excited fluorescence.⁵ The results are shown in Table 1. The fluorescence measurement at $\phi = 1.01$ is the average over four measurements at various heights above the burner, all in the burnt gas region, shown in Table 2. As can be seen, the temperature does not vary, within error limits, above the burner. This result is important since the reversal resolution was only good to about 1 cm in this study. For the fluorescence technique, the resolution was much better, the beam diameter being about 200 microns. The agreement between the results is most gratifying. Note also that the temperature probably peaks near an equivalence ratio of 1.0, as one would expect.

The OH band reversal measurement is essentially a determination of the electronic temperature of the OH in the flame. As such, in spite of the wide acceptance of reversal measurement by the combustion community, the technique is subject to errors as grievous as other less well accepted techniques. Even if great care is taken to avoid systematic errors, as discussed by Snelleman in ref. 6, the electronic temperature of the reversed species can differ from the translational temperature of the flame gases, resulting in error. The technique lacks good spatial resolution. It is a line of sight method which cannot be easily applied in flames with large horizontal temperature gradients. Yet, it is an inexpensive and relatively easily applied technique.

The laser-excited fluorescence measurements were performed in the (1,1) vibrational band of the A $^2\Sigma^+ \leftarrow X ^2\Pi$ electronic transition.⁵ The temperature measured was the rotational temperature in the first excited vibrational state of the ground electronic state of OH. Measurements by this technique could also be subject to effects of nonequilibrium with the translational temperature. However, one would intuitively expect the rotational levels in the ground electronic state to equilibrate much more quickly than electronic states due to the relative energy transfer rates involved. Thus, if a reaction produces electronically excited OH, the slow equilibration could be especially problematic leading to

⁵ W. R. Anderson, "Laser Excited Fluorescence Measurement of OH Rotational Temperatures in a CH₄/N₂O Flame", Eastern Section Meeting, Combustion Institute, Atlanta, Georgia, Nov 1979, paper 3; L. J. Decker and W. R. Anderson, to be published.

⁶ W. Snelleman, "Errors in the Method of Line-Reversal", Combust. Flame 11, 453 (1967).

TABLE 1. CH₄/N₂O TEMPERATURES IN THE BURNT GAS REGION.
THE TEMPERATURES WERE MEASURED AT SEVERAL EQUIVALENCE
RATIOS, $\phi = 4[\text{CH}_4]/[\text{N}_2\text{O}]$.

<u>ϕ</u>	<u>Line Reversal [K]</u>	<u>Laser-Excited Fluorescence [K]</u>
1.18	2129 \pm 50	2113 \pm 38
1.01	2194 \pm 45	2199 \pm 53*
0.55	2089 \pm 70	not measured

*Average of the four measurements in Table 2. The temperature above the burner does not change in the burnt gas region so that this average is meaningful.

TABLE 2. CH₄/N₂O TEMPERATURE AS MEASURED BY LASER EXCITED FLUORESCENCE. THE TEMPERATURE IS MEASURED AS A FUNCTION OF HEIGHT ABOVE THE BURNER AT THE EQUIVALENCE RATIO $\phi = 1.01$

<u>Height (cm)</u>	<u>Temperature (K)</u>
0.25 \pm 0.05	2220 \pm 48
0.50 \pm 0.05	2186 \pm 55
1.00 \pm 0.05	2201 \pm 76
2.50 \pm 0.05	2189 \pm 36

erroneously high temperature measurements. Equilibration in the burnt gas region of atmospheric pressure flames is not expected to be a serious problem as no reactions leading to electronically excited OH are expected to be important and rotational equilibration is rapid.* The agreement between the rotational and electronic temperatures substantiates these arguments.

The usefulness of the OH band reversal technique is demonstrated in this work. The technique provides a temperature measurement for comparison with results of less-well-accepted techniques. It is hoped that this paper will help others avoid some of the subtle errors encountered in this work.

V. ACKNOWLEDGMENTS

The author would like to thank Mr. Mark A. Dewilde for assistance setting up the burner and Dr. John A. Vanderhoff for help in taking some of the data and for a critical reading of the manuscript.

*In emission spectra of radicals in the primary reaction zones of flames, as discussed in ref 1, the radicals are sometimes extremely rotationally hot. Often, the rotational distribution will have two very distinct temperature regimes for the high vs low rotational levels. This phenomenon doubtless occurs due to production of rotationally hot molecules in an excited electronic state due to a chemical reaction. Preferential quenching of states at one end of the distribution probably leads to the observed dual temperature distributions. However, such effects are observed only in low pressure flames.

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